# EFFECT OF DIFFERENT CONCENTRATIONS OF SODIUM CHLORIDE ON THE CORROSION BEHAVIOR OF PURE TITANIUM AND TITANIUM ALLOY COUPLED WITH GOLD ALLOY AND NICKEL CHROMIUM DENTAL CASTING ALLOY

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## CERTIFICATE

This is to certify that this dissertation "**Effect of different concentrations of sodium chloride on the corrosion behavior of pure titanium and titanium alloy coupled with gold alloy and nickel chromium dental casting alloy"** is a genuine work done by **Dr. S. Madhu Mahadevan** under my guidance during his postgraduate study period between 20010 – 2013.

This Dissertation is submitted to THE TAMILNADU Dr. M.G.R. MEDICAL UNIVERSITY, in partial fulfillment for the Degree of **MASTER OF DENTAL SURGERY IN PROSTHODONTICS & CROWN & BRIDGE - BRANCH I.** It has not been submitted (partial or full) for the award of any other degree or diploma.

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## ABSTRACT

**Title:** Effect of different concentrations of sodium chloride on the corrosion behavior of pure titanium and titanium alloy coupled with gold alloy and nickel chromium dental casting alloy

**Aim :** To find out the galvanic corrosion behavior of pure titanium and titanium alloy coupled with gold alloy and nickel-chromium alloy in the presence of sodium chloride at 400 and 1000 ppm.

**Materials and methods:** The specimens selected for the study were pure titanium (PT), titanium alloy(TA),gold alloy (GA) and nickel-chromium alloy (NC). The specimens of dimension 10 x 10 x 2mm was prepared.The test solution used was sodium chloride at 400 ppm and 1000 ppm. A total No of 48 specimens were used. For each system, 6 specimens were studied. The Galvanic corrosion behavior of PT and TA coupled with GA and NC alloy were studied by corrosion test [Potentiostat/Galvanostat ], surface analysis [optical microscope] and elemental release analysis [inductively coupled plasma –mass spectrometry]

**Result:** PT coupled with GA were superior at both 400 and 1000 ppm of sodium chloride in corrosion test ,less pitting corrosion in surface analysis test and less elemental release compared to any other combinations.

**Conclusion:** PT coupled with GA were more corrosion resistance compared to any other combinations at both 400 and 1000 ppm of sodium chloride.

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# *Introduction*

# INTRODUCTION

Commercially pure (Cp) titanium and Ti–6Al–4V alloy have been extensively used as biomedical implants in the recent decades. Although they have an excellent reputation for corrosion resistance and biocompatibility, considerable controversy has been raised over their long - term performance. This is mainly due to the release of titanium , aluminum or vanadium from Ti–6Al–4V alloy and their deposition in tissues , organs and body fluids. Vanadium contained in this alloy has been associated with potential cytotoxic effects and adverse tissue reactions. Further, aluminum present in the alloy was found to produce potential neurological disorders. In addition , long - term experience indicates that the high moduli titanium implants transfer insufficient load to adjacent remodeling bone and this results in bone resorption and eventual loosening of the prosthetic devices.

Attempts were made in the recent past to develop titanium alloys of different compositions to achieve better performance in terms of biomechanical compatibility ( by reducing the modulus ) and bio-chemical compatibility ( by excluding non - toxic elements ). In spite of the better mechanical properties of the presently used

surgical implants , deleterious corrosion process has been reported in certain clinical settings . Corrosion of the implant by the reaction of the body fluid and tissues seems to affect the fatigue life and ultimate strength of the material leading to the mechanical failure of the implants . Further , the presence of particulate corrosion and wear products in the tissues surrounding the implant may ultimately result in a cascade of events leading to bone loss . Hence protection of an implant material from corrosion is indispensable . Titanium and its alloys are found to offer superior corrosion resistance in several media including the human body environment . This high corrosion resistance has been attributed to the formation of a thermodynamically stable TiO2 oxide layer on the surface . However, corrosion resistance of Cp Ti is found to vary significantly with heat treatment temperatures .

More commonly used implant superstructures are gold alloy and nickel-chromium alloy. Concerning metals , most of the allergic reactions are caused by nickel . Nickel-chromium alloys containing 60-80 weight percentage nickel have been developed as an alternative to more expensive gold alloys[5] . The contact of dissimilar metals can create an electrolytic cell and cause galvanic

corrosion . Sodium chloride ( table salt ) is used as a flavoring agent in the cuisines of many cultures . Despite health advisories , maintaining a no - salt or reduced - salt diet has proven difficult for many . It is widely believed that salt intensifies the desirable flavors in food . Another view holds that the value of salt is in its ability to mask undesirable flavors and in this way , the food taste better.

The main sources of sodium intake (excluding table salt) are cereals and cereal products (particularly bread ) , meat and meat products , dry fishes , pickles and even in carbonated beverages . Crisps and savoury snacks tend to be high in salt and can make a significant contribution to sodium intakes . Sea water contains around 35000 ppm of sodium chloride. Nowadays sodium chloride is a constituent of many commercially available toothpaste. Electrolytes in saliva contains sodium and chloride. Sodium is an essential mineral for regulating body fluid balance . Sodium is the most abundant cation in the extracellular fluid and sodium salts account for more than 90% of the osmotically active solute in the plasma and interstitial fluid. Consequently ,sodium load is the major determinant of extracellular volume . Chloride is also important in maintaining the fluid balance and is an essential component of the gastric and intestinal secretions. Sodium is the principal cation of the plasma .

The aim of this study was to find out the effect of sodium chloride on the corrosion behavior , surface roughness and elemental release of pure titanium and titanium alloy when they are coupled with gold alloy and nickel - chromium alloy .

# *Aims and Objectives*

# AIMS AND OBJECTIVES

**This study is undertaken with the following Aims and Objectives.**

To find out the galvanic corrosion behavior of pure titanium and titanium alloy coupled with gold alloy and nickel-chromium alloy in the presence of sodium chloride at 400 and 1000 ppm.

# *Review of Literature*

## REVIEW OF LITERATURE

**Jean Marc Meyer 1977**[1] studied the corrosion resistance of twelve nickel-chromium dental alloys using potentiodynamic polarization curves, polarization resistance curves and potential time curves . He concluded that corrosion resistant Ni-Cr dental alloys result from an accurately balanced composition not only chromium as a passivating element, but also readily oxidized elements like Mo & Mn. The tendency to passivation is enhanced by these two elements along with the resistance to pitting in a chloride containing electrolyte . Their presence markedly increased the passivation and reduced the activity of such alloys and thus reduced the risk of galvanic problems when precious alloys are present in the oral cavity .

**G.J.Ewers et al 1981**[2] studied the corrosion of dental alloys in oral environment. Various dental alloys were used for testing which includes chrome-cobalt alloy, gold alloy, amalgam, gold plated stainless steel pins, silver cones, brass screw post . They concluded that Cobalt-Chromium alloy was very stable in the oral environment but had little ability to passivate at higher voltages . Dental gold alloys behaved predictably and the stainless steel pins, brass screw posts & silver cones revealed corrosion tendencies .

**B.Kasemo et al 1983**[3] studied the surface science and biocompatibility of titanium implant . He stated that the biologic processes elicited by the introduction of an inorganic implant originate in chemical interactions that take place at the interface between the implant and host tissues . When metallic implants are used, the chemical interaction is determined by the properties of a surface oxide and not by the chemical properties of the metal. Contributing factors to the successful results with pure titanium as Implant material may be a combination of the chemical inertness of its oxide , the high dielectric constant of the oxide and some other specific biochemical properties of the oxide.

**Holland R.I. et al 1986**[4] studied the structure & corrosion behavior of low gold alloy . Potentiodynamic and potentiostatic polarization tests were conducted for four different structural state of gold alloy from single phased to multiple phases . They concluded that single phased gold alloys were more corrosive but cause low tarnish than gold alloys with multi- phases which showed more tarnish and less corrosion.

**J.Geis-Gerstorfer et al 1987**[5] carried out invitro investigations of four nickel-chromium alloy in sodium chloride and lactic acid solutions . Electrochemical techniques like potentiodynamic potential curves and Potential time curves were used to analyse the characteristics of these alloys . Substance loss was measured analytically to determine the kind and quantity of de-alloying elements. He concluded that the nickel –chromium alloys containing small amount of Chromium and molybdenum yielded worst results and showed susceptibility to pitting corrosion .

**Robert D. Meyer et al 1993**[6] reviewed the dental history of a patient with symptomatic electrochemical reactions following the occlusal relationship of an existing complete gold crown and silver amalgam restoration . He concluded that the electro-mechanical reaction between dissimilar metals intra-orally was a concern when patient complaints of an unpleasant taste or pain . He further concluded to avoid use of metallic elements in opposing arches intra-orally .

**L.Reclaru et al 1994**[7] demonstrated the study of galvanic corrosion between titanium and dental alloys. Ti / gold based alloys, Ti / palladium based alloys and Ti / non-precious alloys were studied .

Results showed that regarding galvanism, Ti / gold based alloy couplings, Ti / palladium based alloy couplings and Ti / non-precious alloy couplings caused negligible galvanism . There was no crevice corrosion in Ti / gold based alloy couplings and Ti / palladium based alloy couplings. But crevice corrosion was noticed in Ti / non-precious alloy couplings.

**Ramakrishna Venugopalan 1998 et al**[8] states that gold, palladium and silver alloys coupled to titanium were found to be least susceptible to galvanic corrosion . Co– Cr–Mo, Ni–Cr and Fe based alloys coupled to titanium were found to be moderately susceptible to galvanic corrosion due to mechanical– electrochemical interaction . Ni–Cr–Be alloy coupled to titanium was found to be highly susceptible to galvanic corrosion.

**Brigitte Grosgogeat et al 1999**[9] evaluated the galvanic corrosion between Ti / Ti-6Al-4V implants and dental alloys . The electrochemical behavior of 7 alloy superstructures with titanium and Ti-6Al-4V implants was investigated by electrochemical means which includes measurements of galvanic currents in galvanic couple , potential in galvanic couple , measurement of galvanic current in the joints and the assessment of the surface structure .

They concluded that the intensity of the corrosion process was low in the case of Ti / dental alloys and Ti-6Al-4V / dental alloys.

**J.E.G. Gonzalez et al 1999**[10] evaluated the influence of alloying elements and the potential on the corrosion resistance of Ti and other Ti based biomedical implant alloys under simulated physiological conditions . Electrochemical impedance spectroscopy was used and the results were compared with those obtained by potentiostatic and potentiodynamic techniques . They concluded that the titanium based alloys exhibited spontaneous passivity and mo ,v ,fe improved the passivity and limited the active corrosion of the beta phase of titanium. Al enrichment of the alpha phase was found to be detrimental to the passivity and corrosion resistance of titanium .

**M. Cortada et al 2000**[11] investigated the corrosion of five materials for implant suprastructures which includes cast titanium, machined titanium, gold alloy, silver-palladium alloy and nickel-chromium alloy. These materials are galvanically coupled to a titanium implant . Various electrochemical parameters were analyzed. The microstructure of the different dental materials was observed by optical and electron microscopy . Inductively coupled plasma-mass spectrometry technique (ICP-MS) was also used to analyse the

amount of metallic ions Released. The cast and machined titanium had the most passive current density at a given potential and nickel-chromium alloy had the most active critical current density values. The high gold content alloys have excellent resistance corrosion . The palladium alloy had a low critical current density due to the presence of gallium but a selective dissolution of copper-rich phases was observed through energy dispersive X-ray analysis.

**J.Karov et al 2001**[12] investigated the galvanic corrosion behavior of three different amalgam brands coupled to two gold alloys and orthodontic brackets . In the resulting galvanic cells , the amalgam coupled to gold were anodic ,exhibiting galvanic current densities about one order of magnitude higher than the uncoupled corrosion current densities . Coupling amalgams to orthodontic brackets resulted in galvanic current densities of the same magnitude as the uncoupled samples . Brushing caused transient increases in galvanic current densities . Brushing of amalgam / bracket couples , caused an anodic peak followed by brief polarization reversal during which the brackets were anodic.

**I.Ramires et al 2001**[13] studied the electrochemical behavior of Ti-Pd and Ti-6Al-4V alloys in 0.9% NaCl solution using single

triangular potential sweep , potentiodynamic polarization curves and electrochemical impedance spectroscopy techniques (EIS). The potentiodynamic response and EIS measurements indicate that the alloying elements have effect on the basic corrosion resistance of titanium in NaCl solution . The corrosion rate of Ti-Pd is mainly controlled by anodic electron transfer reaction involved in the metal electrodissolution process , whereas in Ti-6Al-4V , the kinetics of the electrodissolution reaction is controlled by diffusion of OH- ions toward or the oxidation products away from the reaction sites through pores in the corrosion films.

**Maan Aziz-kerrzo et al 2001**[14] evaluated the corrosion susceptibility of titanium and its alloys in a buffered saline solutions of 0.9% sodium chloride and deionized water using anodic polarization and electrochemical impedence measurements . Results showed that pure titanium and Ti-6Al-4V exhibited high resistance to the onset of localized corrosion resistance but pits were found to initiate at low potentials . The passive films formed on these electrodes were analysed in terms of a dual oxide layer comprising an inner barrier and an outer porous layer . The nature of the porous layer was found to depend on the nature of the alloy and the solution anion species .

**Renata Poljak et al 2002 stated that the**[15] electrochemical corrosion is the most important damaging factor of dental works . Corrosion is the unintentional wearing down of the metal surfaces and damage to the outer and inner layers of their surface caused by exposure to chemical or electrochemical reaction of the surrounding area. Electrolyte is needed for electrochemical reaction. Corrosive resistance is prerequisite for biocompatibility. The corrosion potential of saliva increases as its pH factor decreases and as chloride concentration increases . In most alloy-electrolyte systems, corrosion stops on the very surface by creation of a surface oxide layer which is good protection from further corrosion . He further stated that the two different protective layers are formed in the mouth : an oxide layer and a biofilm .

**Eser Tufekci et al 2002**[16] stated that the biocompatibility of high-palladium alloy restorations has been of some concern due to the release of palladium into the oral environment and sensitivity reactions in patients . He measured the in vitro elemental release from a Pd-Cu-Ga alloy and a Pd-Ga alloy into a corrosion testing medium containing aqueous lactic acid and NaCl solutionl in 300 mL of ultrapure deionized water . Elemental compositions of the

solutions were analyzed with inductively coupled plasma-mass spectroscopy (ICP-MS). Relative proportions of the elements in the solutions were consistent with the release of palladium and breakdown of microstructural phases found in the alloys . The results of the present study suggest that, of the 2 high-palladium alloys tested, the Pd-Ga alloy may cause fewer problems than the Pd-Cu-Ga alloy interms of patient sensitivity to palladium .

**Nadia M.Taher et al 2003**[17] conducted a study to evaluate the galvanic corrosion behavior of Co-Cr , Ni-Cr, silver-palladium, gold and ternary titanium when coupled with endosseus titanium implant abutment material controls. Results in this study indicate that the gold alloy was an excellent couple with a titanium implant .Silver-palladium alloy and cobalt-chromium alloy when coupled with titanium implant showed acceptable galvanic corrosion behavior. Ni-Cr base metal alloy showed unstable galvanic corrosion behavior when coupled with a titanium implant .Amalgam alloy resulted in higher galvanic interactions when coupled with a titanium implant. Commercially pure titanium when coupled with endosseus titanium implant healing abutment showed unexpected galvanic corrosion behavior .

**Keun-Taek Oh et al 2004**[18] conducted a study to investigate the galvanic and crevice corrosion characteristics in combination of gold, silver–palladium, cobalt–chromium and nickel–chromium suprastructures with titanium implants . Potentiodynamic and potentiostatic testing were performed . After electrochemical testing, surface morphologies and cross-sections were examined using micrographs of the samples. Results showed that the galvanic corrosion occur in the Co–Cr alloy, but this is determined to have low risk. In the other alloys, the possibility of corrosion and its risks appears lower . He further stated that because of the structure of the implant, the risk of the crevice corrosion exists.

**S´ergio Luiz de Assis et al 2006**[19] investigated the electrochemical behavior of Ti–6Al–4V and Ti–6Al–7Nb alloys, commonly used implant materials together with that of Ti–13Nb–13Zr alloy . The aim of present study was to evaluate their corrosion resistance in an artificial physiological solution . This evaluation was carried out through the analysis of the corrosion potential variation with time, potentiodynamic polarization curves and electrochemical impedance spectroscopy (EIS) tests. The electrochemical technique resulted in very low corrosion current densities for the three titanium alloys tested showing that they are passive . The EIS results indicated that

the film formed on the Ti alloys is composed of a bi-layered oxide consisting of an inner barrier layer associated to high impedance and responsible for corrosion protection and an outer porous layer of lower impedance, which apparently facilitates the osseointegration .

**Deepthi Upadhyay et al 2006**[20] reviewed the corrosion of different dental metals and alloys together with oral environments and various types of corrosion . She concluded that the nature of dental metal alloys plays a major role in the initiation and propagation of corrosion. Therefore, as for as possible corrosion free / least corrosivemetal alloys should be used for the artificial tooth and repair of the cavities for good health .

**christopher M.Wylie et al 2007**[21] studied the microstructure and corrosion behaviour of two nickel - based dental casting alloys before and after a heat treatment. The microstructure was studied using scanning electron microscopy ( SEM ) and energy dispersive X - ray analysis ( EDX ). Corrosion behaviour was evaluated by electrochemical measurements in the presence of a crevice. Small changes in microstructure were observed after heat treatment but had a negligible effect on the corrosion properties in the conditions tested . The alloy with a lower bulk level of chromium showed

lower corrosion resistance, indicated by an increased passive current density . Selective dissolution occurred at regions within the microstructure containing lower levels of Cr and Mo . The alloy containing a higher level of chromium showed superior corrosion resistance, which was associated with a more uniform distribution of chromium in the alloy microstructure . He concluded that the presence of crevices combined with an inhomogeneous distribution of chromium in the microstructure can lead to accelerated corrosion of nickel based alloys with lower chromium contents . He further concluded this effect can be avoided by increasing the chromium content of the alloy .

**Mohamed F. Ayad et al 2008**[22] carried out an in vitro study to characterize the elemental composition of an as - received and recast high-noble alloy and to examine the corrosion behavior in 2 media in 0.09% NaCl solution and Fusayama artificial saliva, using a potentiodynamic polarization technique . The elemental map showed decreased composition of Au for the recast group . The bulk composition of Au, Ag and Pd remained constant, but the copper and zinc contents slightly decreased . The results indicate that the high noble alloy tested produced acceptable castings that are corrosion resistant when recast . He concluded that the high noble

alloy in all casting protocols evaluated retained passivity under electrochemical conditions similar to the oral environment .

**T.P.Chaturvedi et al 2009**[23] reviewed the various aspects of corrosion and biocompatibility of dental titanium implants as well as suprastructures . He stated that the biocompatibility of the implants and its associated structures is important for proper functioning of the prosthesis in the mouth . corrosion can severely limit the fatigue life and ultimate strength of the material leading to mechanical failure of dental materials . High noble alloys used in dentistry are so stable chemically that they do not undergo significant corrosion in the oral environment ,the major components being gold ,platinum and palladium. Small galvanic currents associated with electrogalvanism are continuously present in the oral cavity . He concluded that the choice of materials used for implant as well as implant borne suprastructures becomes crucial and can be made by way of evaluating their galvanic corrosion behaviours .

**Pascal De March et al 2009**[24] studied the electrochemical behaviors of alloys for fixed partial dentures . High Noble , Noble alloy ( less rich in precious metals ) and Predominantly Base alloys ( based on nickel and chromium) and post - solder alloys used in dental

prostheses were specified in nacl– **9g/l**-aqueous solution . The galvanic corrosion between each parent alloy and its usual post - solder was also studied. Despite their differences of chemical compositions most of the precious metal - based parent alloys and post - solder alloys cannot be significantly corroded when they are in contact with a neutral NaCl-containing aqueous solution . On the contrary , the less expensive PB alloys , which are protected against corrosion only by a passivation layer are slowly corroded . However , a galvanic corrosion can occur when a parent alloy and its usual post - solder are coupled .

**Suleyman Hakan Tuna et al 2009**[25] studied the electrochemical properties of four dental casting alloys coupled with titanium implants. The potentiodynamic polarization curves and open – circuit potentials (OCP) of four UCLA type suprastructures coupled with implant fixtures were taken in Afnor type artificial saliva solution with nacl of 0.7g/l at 37°C . The concentration of ions leached was estimated with ICP - MS. He concluded that the titanium implants rapidly formed a stable, passive layer .In the titanium implant and suprastructure alloy combinations, the titanium implant had virtually no effect on the pitting potential due to the passive layer formed on the surface of the implant. The corrosion properties were highly

related to the suprastructure alloys . The gold and palladium-based alloys were found to have dissolved less than the nickel-chromium and cobalt-chromium alloys .

**Rahul Bhola et al 2009**[26] studied the electrochemical behavior of titanium and its alloy as dental implants in normal saline with 2.22g of sodium chloride . He concluded that in normal saline solutions, all alloys exhibit high corrosion resistance and the corrosion rates observed fall in the acceptable range for biocompatibility of metallic implants . All alloys show considerable ennoblement and form a stable titanium oxide film as indicated by their pH and potential position in the pourbiax diagram . From cyclic polarization curves obtained it was concluded that the alloys resist localized corrosion .

**D. Mareci et al 2010**[27] studied the electrochemical determination of the Corrosion resistance of four NiCr Alloys (Wirolloy, Heraenium, Nicromal Soft and VeraSoft ) based on the polarization curves and electrochemical impedance spectroscopy maintained in 0.1M NaCl solution . He concluded that very low corrosion current densities, typical of Passive materials, were obtained for all the samples tested in 0.1M NaCl solutions . For VeraSoft and Nicromal Soft alloys, low

and Dangerous breakdown potential values may be recorded . Over the Surface of the Heraenium alloy a uniform corrosion appears, while inCase of the Wirolloy, VeraSoft and Nicromal Soft alloys surfacea pitting corrosion is developed . Heraenium alloy with high Cr and Mo content exhibit a much wider passivation range and a better resistance to pitting corrosion . The EIS results show that Ni-Cr based alloys exhibit passivity at open circuit potential . The Heraenium and Wirolloy alloys with dendrites microstructure show the best electrochemical behaviour in 0.1M NaCl solutions .

**Geetha Manivasagam et al 2010**[28] reviewed the Corrosion and its Prevention of biomedical implants **.** Cobalt-chromium alloy which is a commonly used biomaterial consists of the elements cobalt, chromium, nickel and molybdenum . It is felt that the corrosion of cobalt-chrome in the wet and salty surroundings of the human body, releases toxins into the body which in turn leads to the formation of cancerous tumors and they could exist at other parts of the human body due to the released ions . She further concluded that the biomedical materials which are subjected to cyclic loading and high stresses in the presence of aggressive environment fail due to fatigue . During fatigue there is disruption of the oxide layer and the inability of the material to repassivate immediately exposes some

region of the metal to the environment leading to corrosion . Surface modifications are often performed on the biomedical implants to improve corrosion resistance, wear resistance, surface texture and biocompatibility .

**M.Atapour et al 2010**[29] studied the corrosion behavior of four different Ti-6Al-4V with different thermo mechanical treatments and microstructures investigated in sodium chloride ( NaCl ) and hydrochloric acid ( HCl ) solutions . The microstructure constituent morphologies and distributions were characterized with optical and scanning electron microscopy while constituent compositions were determined with energy-dispersive x-ray analysis . All four microstructures exhibited spontaneous passivity in 0.9% NaCl at and active/passive transitions in 1.5 M HCl . Very little difference in corrosion rate was observed using potentiodynamic polarization curves and weight-loss tests . All microstructures exhibited spontaneous passivity and similar potentiodynamic curves in 0.9% NaCl solution and active to passive transitions in 1.5 M HCl solution .

**Z. Paszenda et al 2010**[30] analysed the surface roughness impact and the influence of the steam sterilisation process on

physicochemical properties of samples made of Ti-6Al-4V and Ti-6Al-7Nb alloys after their exposure in a solution simulating the osseous environment . Potentiodynamic tests, which were carried out in an environment simulating the osseous system of humans, revealed a diversified corrosion resistance of selected biomaterials on the basis of the electrochemical studies . A favourable influence of steam sterilization on corrosion resistance of Ti-6Al-4V and Ti-6Al-7Nb alloys, regardless of the applied mechanical treatment was observed. Exposition to the solution simulating human osseous system environment did not influence over physicochemical properties of the passive layer.

**Rabab M. Abou Shahba et al 2011**[31] studied the corrosion and inhibition of Ti-6Al-4V Alloy in NaCl Solution . The electrochemical behavior of Ti-6Al-4V alloy has been studied in NaCl solutions using Anodic and cathodic potentiodynamic polarization measurements and cyclic polarization measurements . From the results of open-circuit potential measurements of Ti-6Al-4V, a general tendency for the immersion potential to shift steadily towards nobler values indicated formation of a passive layer of TiO2 and additional metal oxides . It was also found that Ecorr was shifted to more positive values and that Icorr increased with

increasing NaCl concentration . Potentiodynamic cyclic anodic polarization measurements for Ti-6Al-4V alloy in NaCl solutions showed active, passive and trans-passive regions, indicating that the alloy was susceptible to pitting corrosion.

**Rahul Bhola et al 2011**[32] reviewed the corrosion in titanium dental implants/prostheses . The metallic titanium dental implants/prostheses derive their biocompatibility from the alloying elements responsible for the formation of a continuous stable TiO2 passive film on its surface . There is a significantly small release of alloying ions even under the ideal conditions of passivity and with no damage to the implant surface . Corrosion of these implants may occur when the oral conditions are unfavorable as under mechanical trauma to the implant surface or the use of inappropriate metal combination as auxiliary prostheses (galvanism).

**B. B. Zhang et al 2011**[33] studied the corrosion behaviour of newly developed Ti–Ag–Fe dental alloys in neutral saline solution . The corrosion behavior of Ti–5Ag–Fe alloys in neutral saline solution was investigated by the open-circuit potential, potentiodynamic polarization, electrochemical impedance spectroscopy and potentiostatic tests . The microstructural observation indicated that b-

Ti phase was retained by the addition of Fe into Ti–Ag alloys . Compared with commercially pure (CP) Ti, Ti–5Ag–Fe alloys exhibited higher corrosion potentials, lower current densities and larger impedance, these suggested that Ti–5Ag–Fe alloys have nobler electrochemical corrosion behavior when compared with CP Ti in neutral saline solution.

# *Materials And Methods*

# MATERIALS AND METHODS

The specimens selected for the study were pure titanium (PT), titanium alloy(TA),gold alloy (GA) and nickel-chromium alloy (NC). The composition and manufacturer of materials were shown in Table – I. The specimens of dimension 10 x 10 x 2mm was cut from parent metal, smoothened with silicon carbide paper No. 400 down to No.120 in series, degreased with trichloroethylene, followed by deionized water.

The test solution (electrolyte) used was sodium chloride [NaF] (Loba chemie, India) at 400 ppm and 1000 ppm. The systems used in this study were listed in Table II. A total No. of 48 specimens were used. For each system, 6 specimens were studied.

## GALVANIC CORROSION STUDY

The Galvanic corrosion behavior of PT and TA coupled with GA and NC alloy were studied by corrosion test, surface analysis and elemental release analysis. The reference electrode used was saturated calomel electrode and platinum electrode was used as a counter electrode. The reference electrode, counter electrode and the specimens were immersed in 100 ml of test solution contained in a 250ml beaker. The current was passed at a scanning rate of 1800mV/hr. Changes in

corrosion potential with immersion time were measured in the test solution for 60 minutes [Potentiostat/Galvanostat,Voltalab21(PGP201and Voltamaster4),Radiometer analytical SAS,France]. Then the electrodes were removed from the test solution, gently rinsed with distilled water and air dried. The surface analysis test was done by examining the surfaces of specimens with optical microscope (Eclipse LV100D, Nikon, USA) to study the corrosion behavior. The elemental release analysis test was carried out by quantitatively analyzing the elements (metal ions) released in the test solution from the galvanic coupling alloys by employing inductively coupled plasma –mass spectrometry (Plasma Quad ll, V.G. Elemental, Canada). Results were statically analyzed by one way ANOVA test.

## TABLE – I

| Material | Composition | Supplier |
|---|---|---|
| Pure Titanium | 99.2 % metal basis | ALFA AESAR, U.S.A. |
| Ti6AL4V alloy | Ti: base, Al : 6, V:4 | SOUTH ASIA METALS PVT LTD., India |
| Ni-Cr alloy | Ni -61.4, Cr – 25.7, Mo-11, Si – 1.5, Mn <1, Al <1, C<1 | IVOCLAR VIVADENT, Switzerland |
| Gold Alloy MaxiGold | Au – 59.5, Ag – 26.3, Pd – 2.7, Cu – 8.5, Zn – 2.7, In <1, Ir<1 | IVOCLAR VIVADENT, Switzerland |

## TABLE – II

| S.No | Electrode | System |
|---|---|---|
| 1 | Pure titanium with gold alloy | PT – $GA_1$ → 400 ppm NaCl<br>PT – $GA_2$ → 1000 ppm NaCl |
| 2 | Pure titanium with nickel-chromium alloy | PT – $NC_1$ → 400 ppm NaCl<br>PT – $NC_2$ → 1000 ppm NaCl |
| .3 | Ti6Al4V alloy with gold alloy | TA – $GA_1$ → 400 ppmNaCl<br>TA – $GA_2$ → 1000 ppm NaCl |
| 4 | Ti6Al4V with nickel-chromium alloy | TA – $NC_1$ → 400ppm NaCl<br>TA – $NC_2$ → 1000 ppm NaCl |

PT - Pure titanium

TA - Titanium alloy (Ti6Al4V)

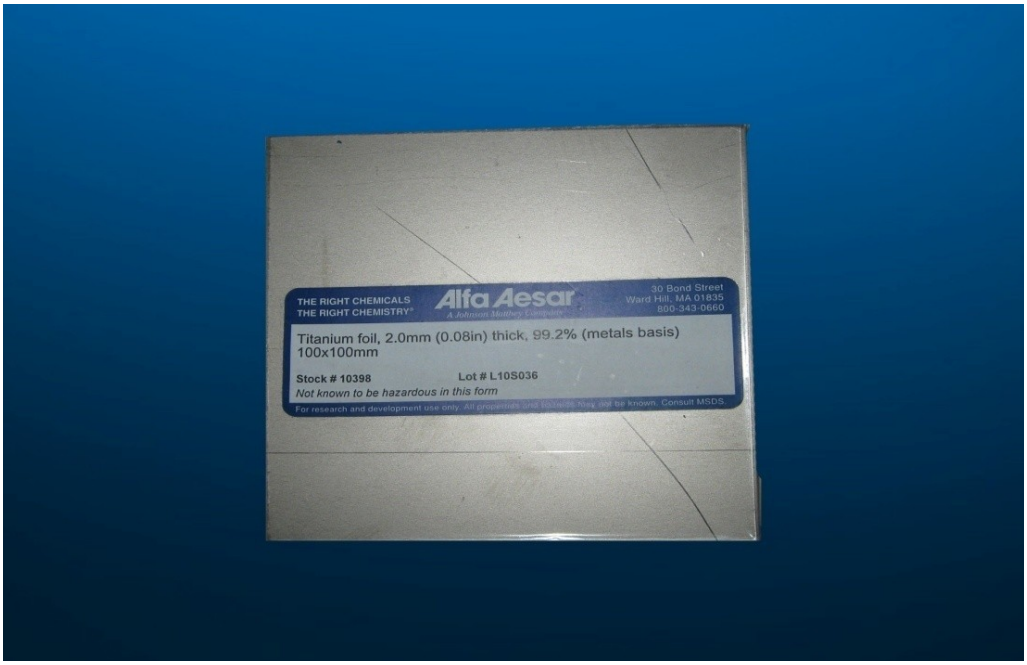
NC - Nickel chromium alloy

GA - Gold alloy

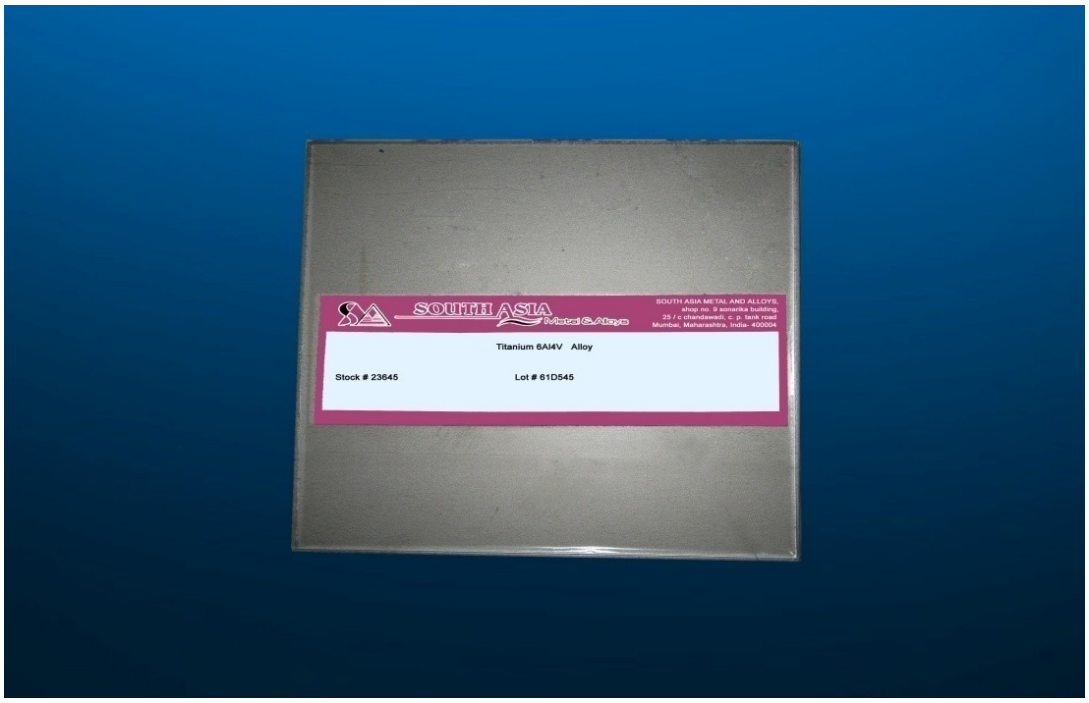
NaCl - Sodium chloride

**Colour Plate – I**

**MATERIALS USED IN THIS STUDY**




**PURE TITANIUM**

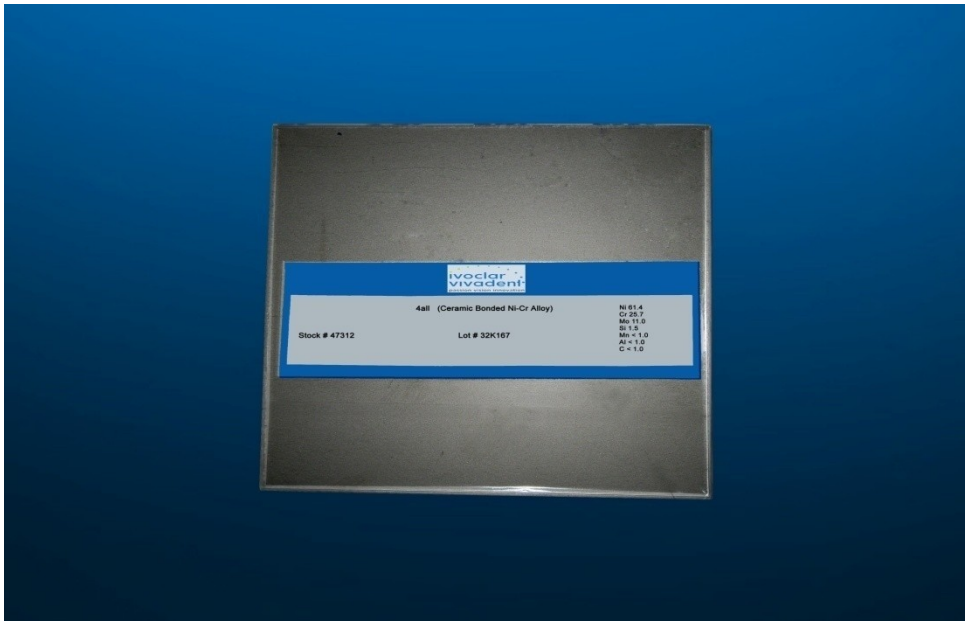



**Ti6Al4V ALLOY**

**Colour Plate – II**

**MATERIALS USED IN THIS STUDY**

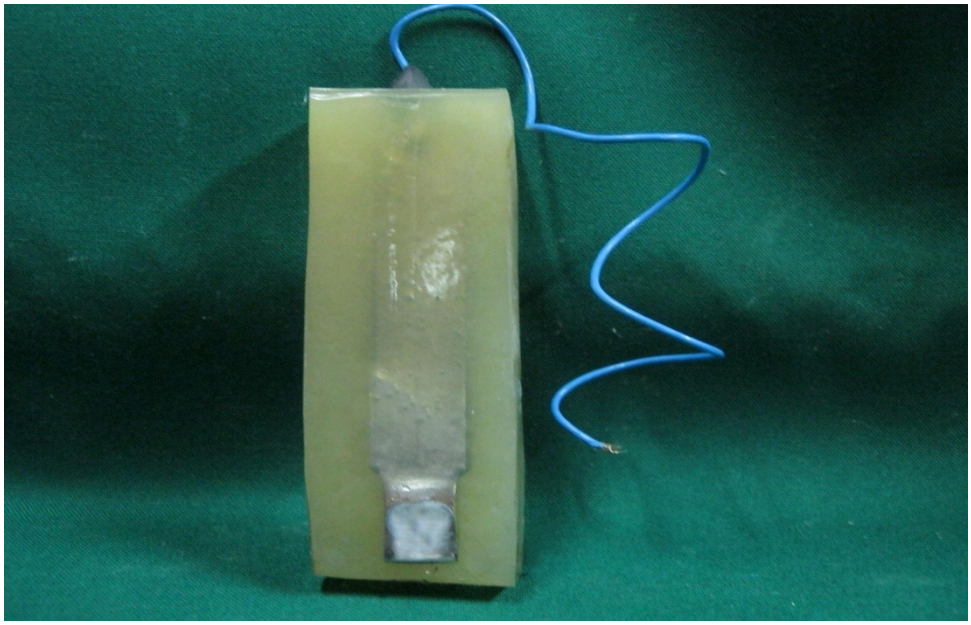

**GOLD ALLOY**



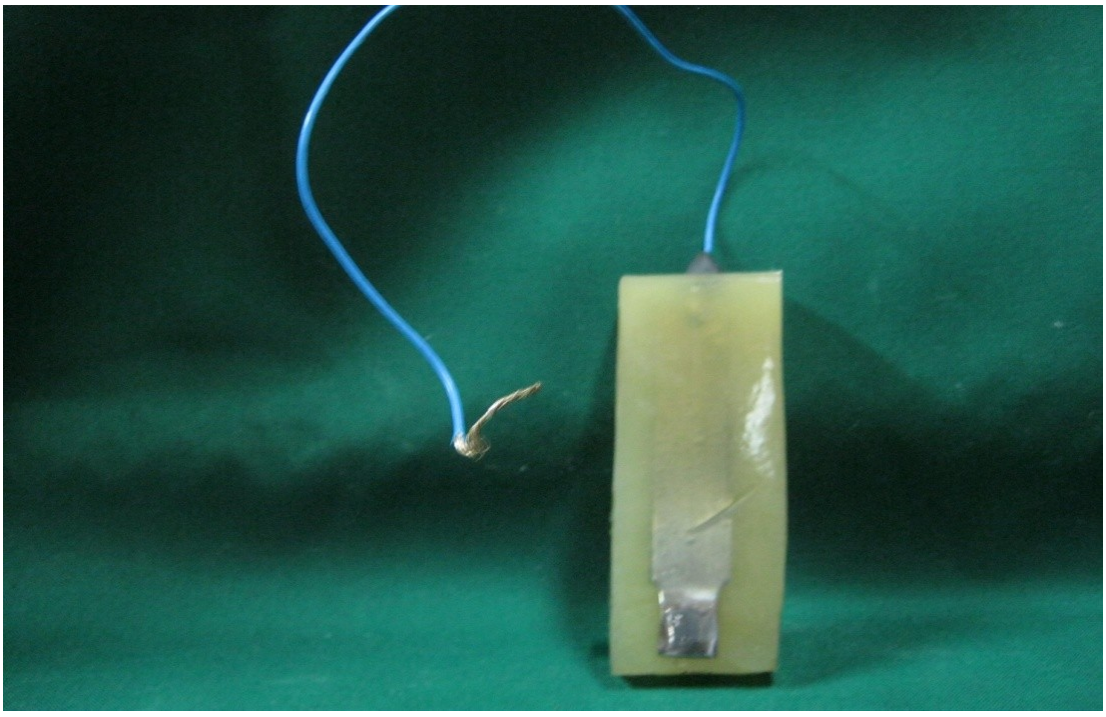

**NICKEL - CHROMIUM ALLOY**

**Colour Plate – III**

**SPECIMEN USED IN THIS STUDY**



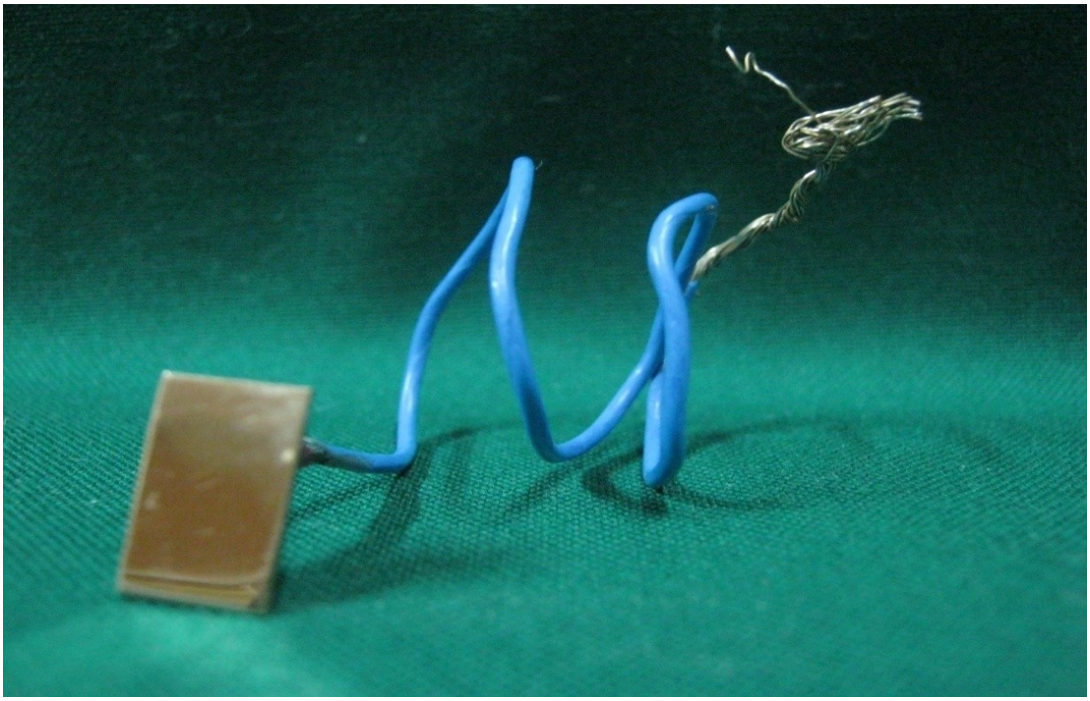
**PURE TITANIUM**



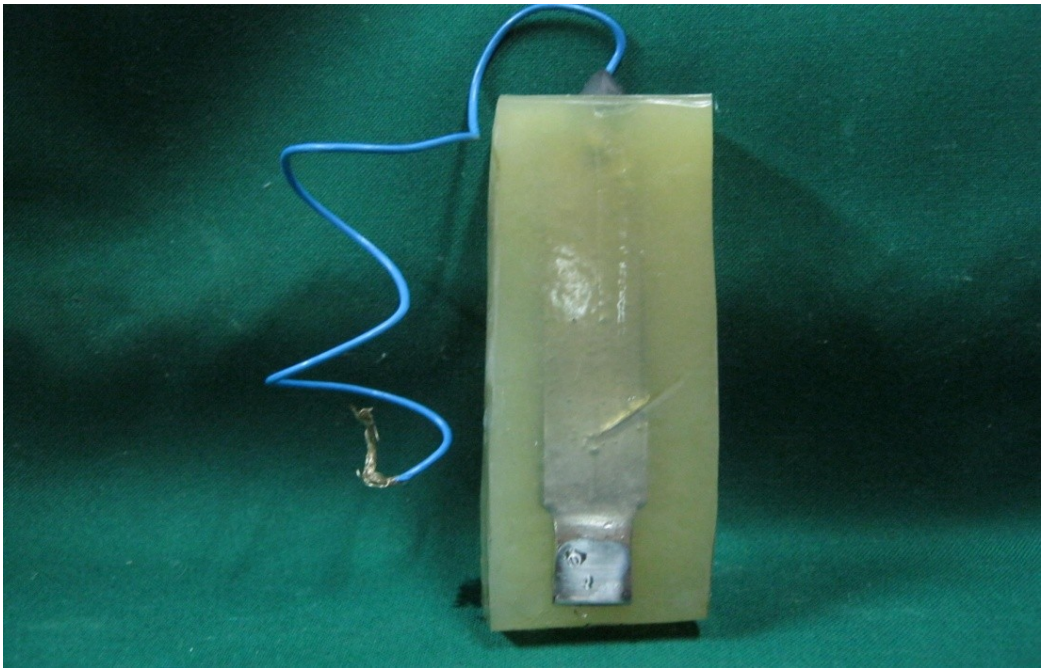
**Ti6Al4V ALLOY**

**Colour Plate – IV**

**SPECIMEN USED IN THE STUDY**



**GOLD ALLOY**



**NICKEL - CHROMIUM ALLOY**

**Colour Plate – V**

**POTENTIOSTAT / GALVANOSTAT**

**Colour Plate – VI**

**OPTICAL MICROSCOPE**

**Colour Plate – VII**

**INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRY (ICPMS)**

# *Results and Observations*

# RESULTS AND OBSERVATIONS

## CORROSION TEST RESULTS

The galvanic coupling of PT and TA with GA and NC alloy in the presence of sodium chloride at 400 and 1000 ppm were analysed by potentiostat. PT was more stable than TA when coupled with GA at 400 ppm of NaCl. PT and TA when coupled with NC showed a very low potential indicating a significant amount of corrosion in the presence of NaCl at 400 ppm and more corrosive at 1000 ppm of NaCl. PT was superior to TA when coupled with GA at 1000 ppm of NaCl. Results showed that PT coupled with GA was superior to any other combinations at 400 and 1000 ppm of NaCl.

## SURFACE ANALYSIS RESULTS

The metal surface of PT and TA coupled with GA and NC alloy were analysed with optical microscope.The metal surfaces of PT were superior to TA when coupled with GA at 400 ppm and 1000 ppm of NaCl.PT and TA when coupled with NC showed pitting corrosion of NC at 400 ppm and more pits were noticed at 1000 ppm of NaCl.The surface of PT did not show any more roughening in the presence of NaCl but TA showed a slight increase in roughness when coupled with GA and NC alloy at 400 ppm and 1000 ppm. The optical microscopic results showed

that PT coupled with GA were superior in corrosion resistance at 400 and 1000 ppm of NaCl when compared with any other combinations.

## ELEMENTAL RELEASE RESULTS

The Amount of elements released in the test solution were analysed using inductively coupled plasma mass spectrometry. PT and TA when coupled with GA showed negligible amount of elements released at 400 ppm and 1000 ppm of NaCl. When NC alloy was coupled with PT and TA the amount of NC alloy released in the test solution was more at 1000 ppm when compared to 400 ppm of NaCl indicating a significant amount of corrosion. In the presence of NaCl at 400 and 1000 ppm, the TA release was more compared to PT when coupled with both GA and NC alloy. The mean value of TA coupled with NC alloy at 400 ppm was 14 for nickel, 6 for chromium, 4 for TA, whereas at 1000 ppm it was 22 for nickel, 10 for chromium, 8 for TA. This combination shows more elements released in the test solution than any other combinations. Thus the elemental release results showed that PT coupled with GA showed very less amount of elemental release at 400 and 1000 ppm of NaCl compared to any other combinations.

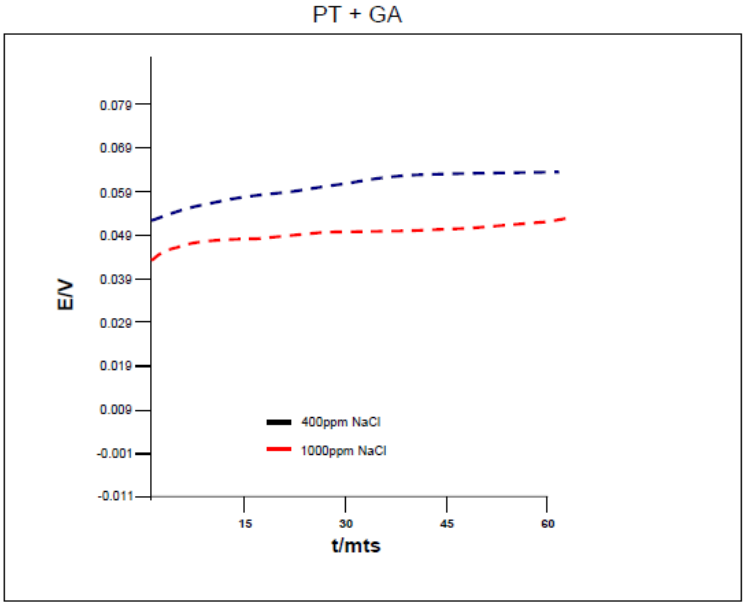



Fig. 1. shows the Galvanic Potential of PT coupled with GA in the presence of NaCl at 400 and 1000 ppm

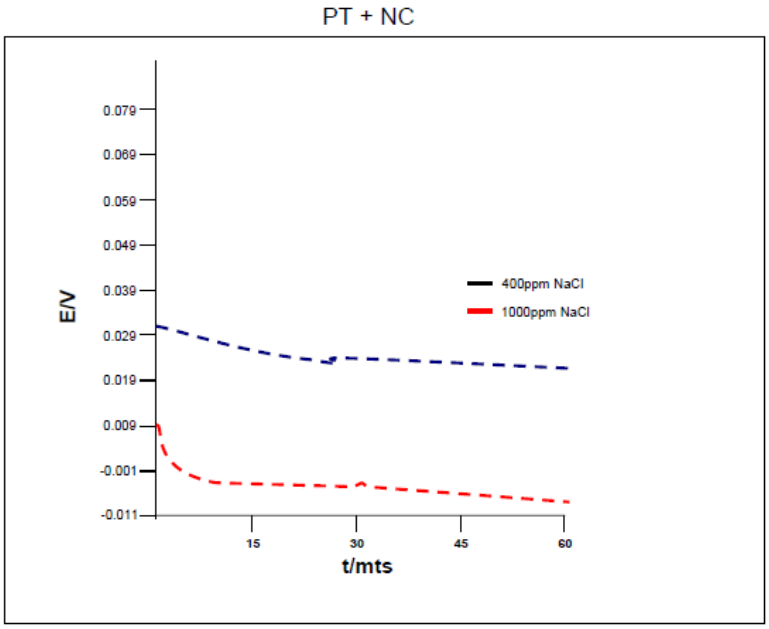



Fig. 2. shows the Galvanic Potential of PT coupled with NC in the presence of NaCl at 400 and 1000 ppm

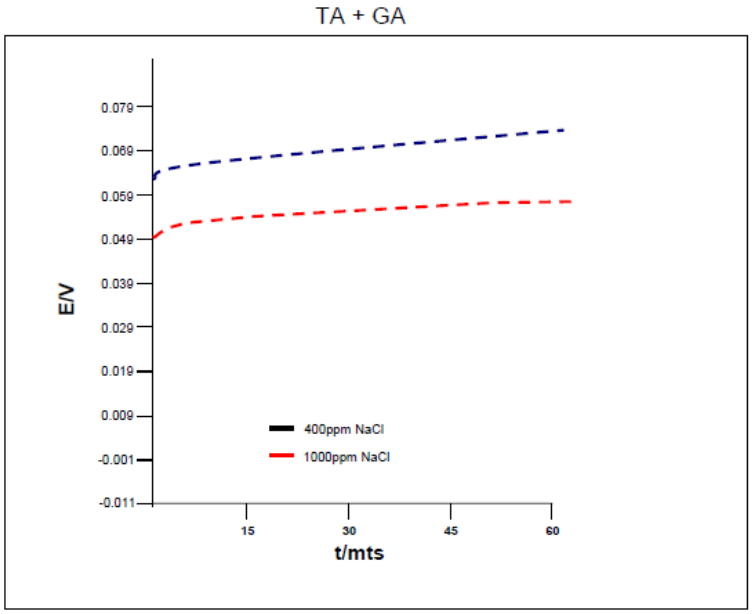



Fig. 3. shows the Galvanic Potential of TA coupled with GA in the presence of NaCl at 400 and 1000 ppm

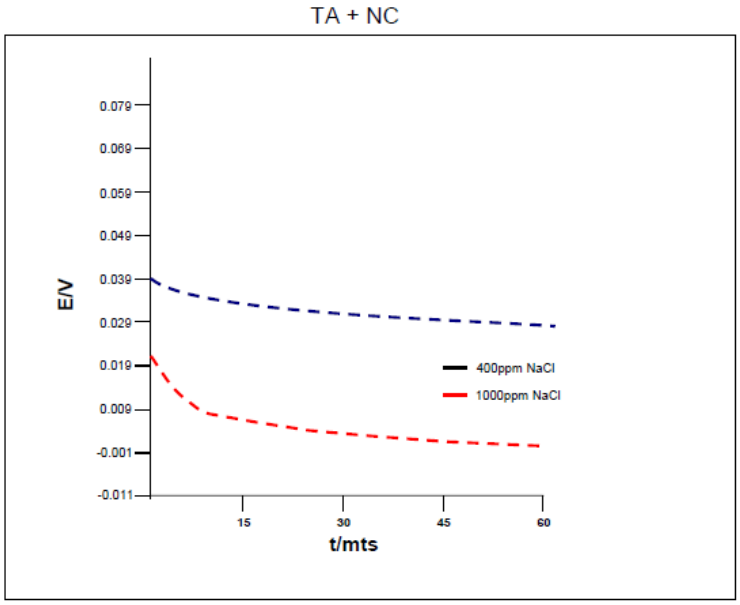



Fig. 4. shows the Galvanic Potential of TA coupled with NC in the presence of NaCl at 400 and 1000 ppm

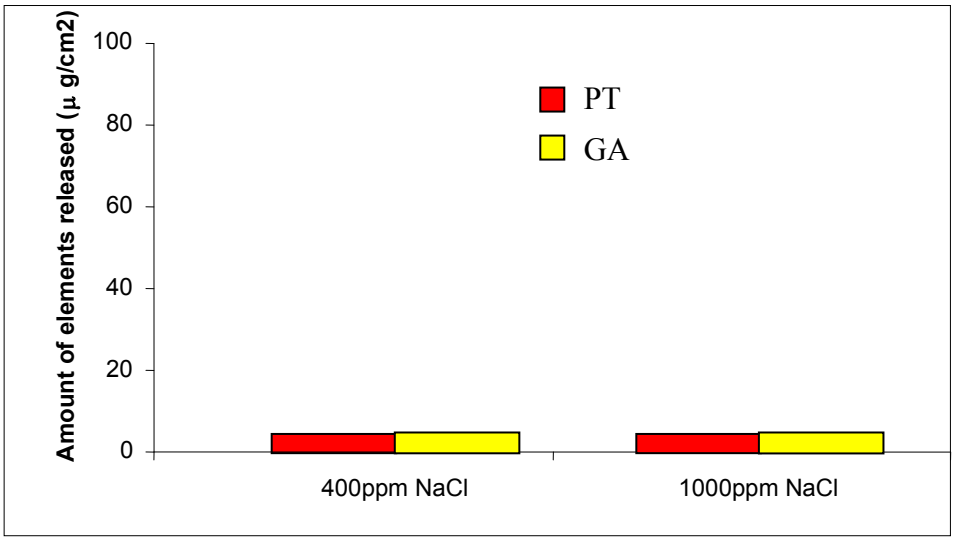



Fig.5. shows the amount of elements released when PT coupled with GA in the presence of NaCl at 400 and 1000ppm

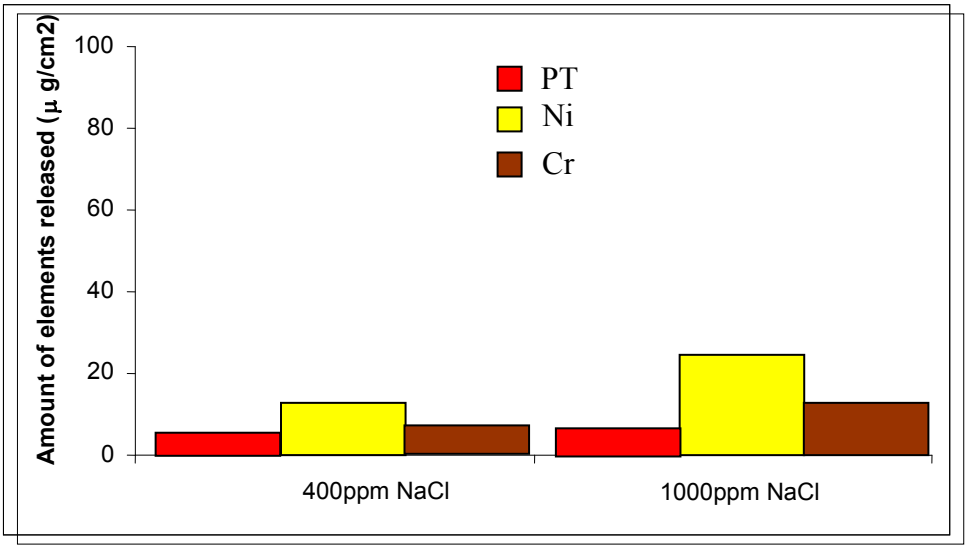



Fig .6 shows the amount of elements released when PT coupled with NC in the presence of NaCl at 400 and 1000ppm

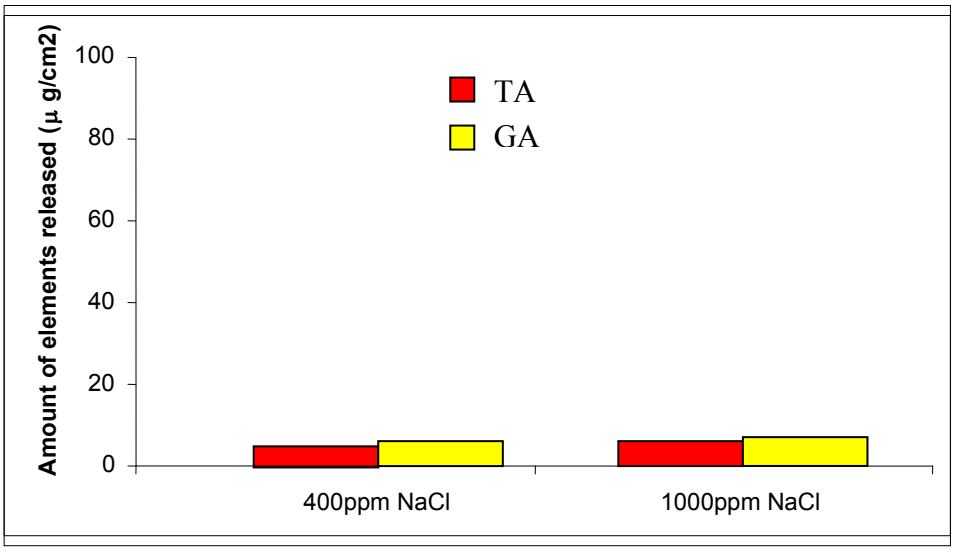



Fig .7 shows the amount of elements released when TA coupled with GA in the presence of NaCl at 400 and 1000 ppm

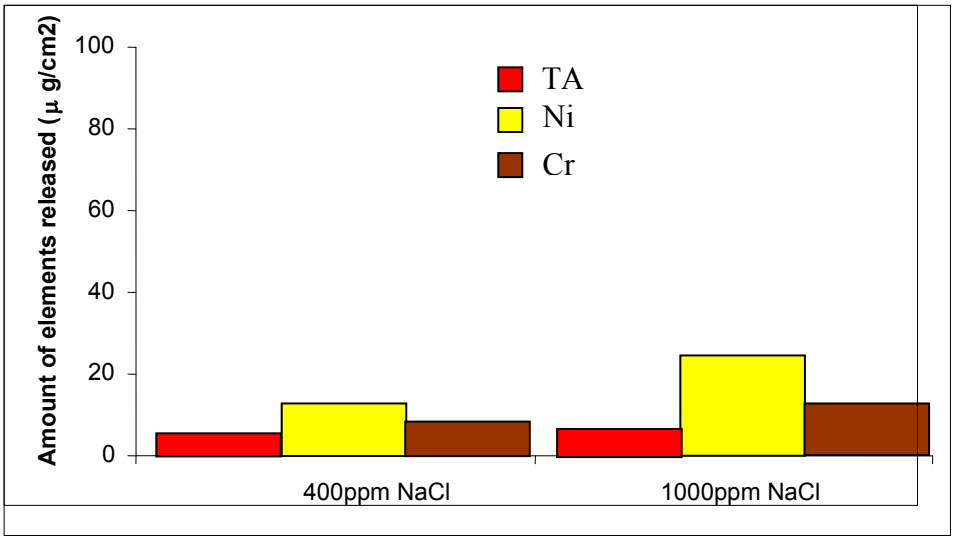



Fig 8 shows the amount of elements released when NC coupled with TA in the presence of NaCl at 400 and 1000ppm

FIG. 9 OPTICAL MICROSCOPIC PICTURE OF METALS AFTER GALVANIC CORROSION

| **NaCl Concentration** | **METALS** | |
|---|---|---|
| | PT | GA |
| 400ppm |  |  |
| 1000 ppm |  |  |

FIG. 10 OPTICAL MICROSCOPIC PICTURE OF METALS AFTER GALVANIC CORROSION

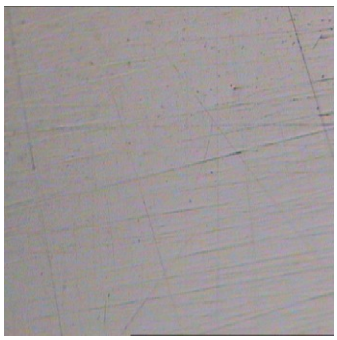
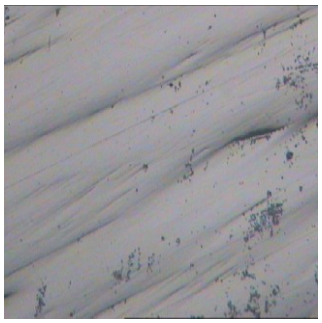
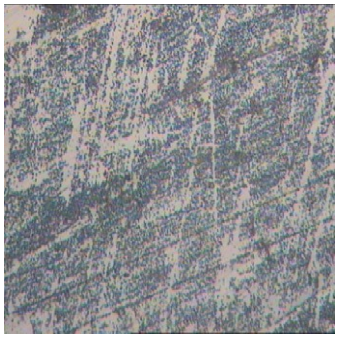
| **NaCl Concentration** | **METALS** | |
|---|---|---|
| | PT | NC |
| 400ppm |  |  |
| 1000 ppm |  |  |

FIG. 11 OPTICAL MICROSCOPIC PICTURE OF METALS AFTER GALVANIC CORROSION

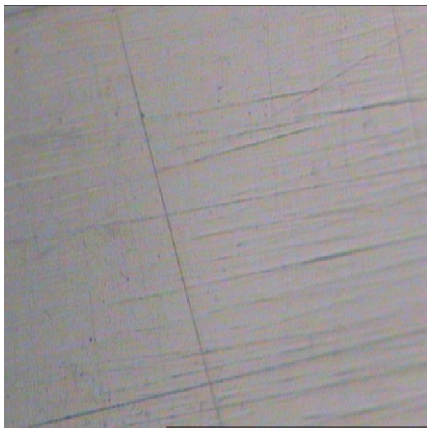
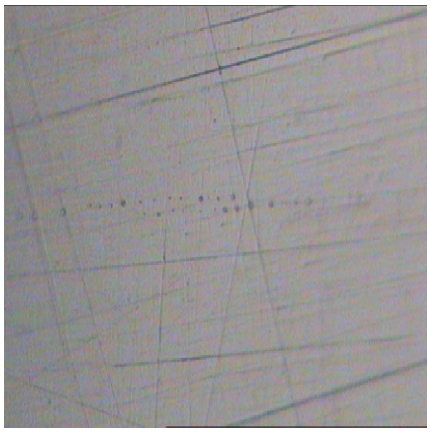
| **NaCl Concentration** | **METALS** | |
|---|---|---|
| | TA | GA |
| 400ppm |  |  |
| 1000 ppm |  |  |

FIG. 12. OPTICAL MICROSCOPIC PICTURE OF METALS AFTER GALVANIC CORROSION

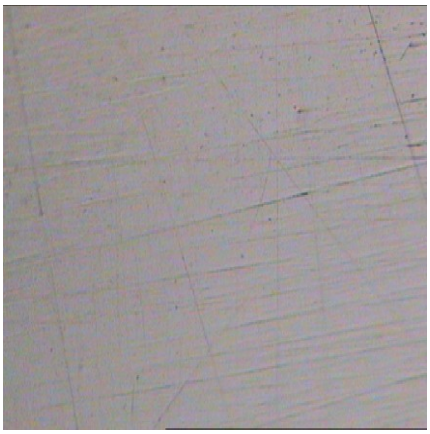
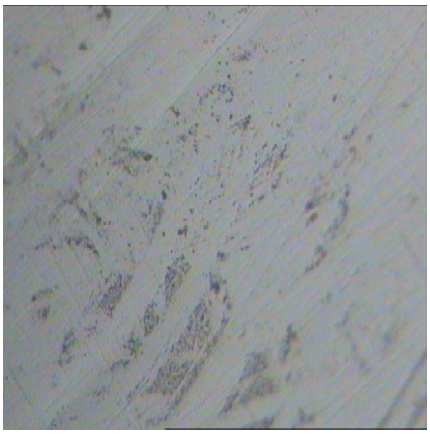
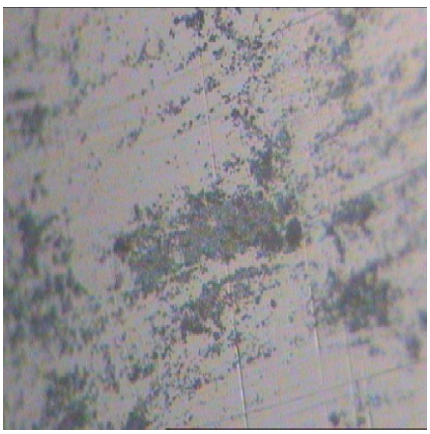
| **NaCl Concentration** | **METALS** | |
|---|---|---|
| | TA | NC |
| 400ppm |  |  |
| 1000 ppm |  |  |

**Fig 13 Statistical analysis of elemental release graph when PT Coupled with GA**

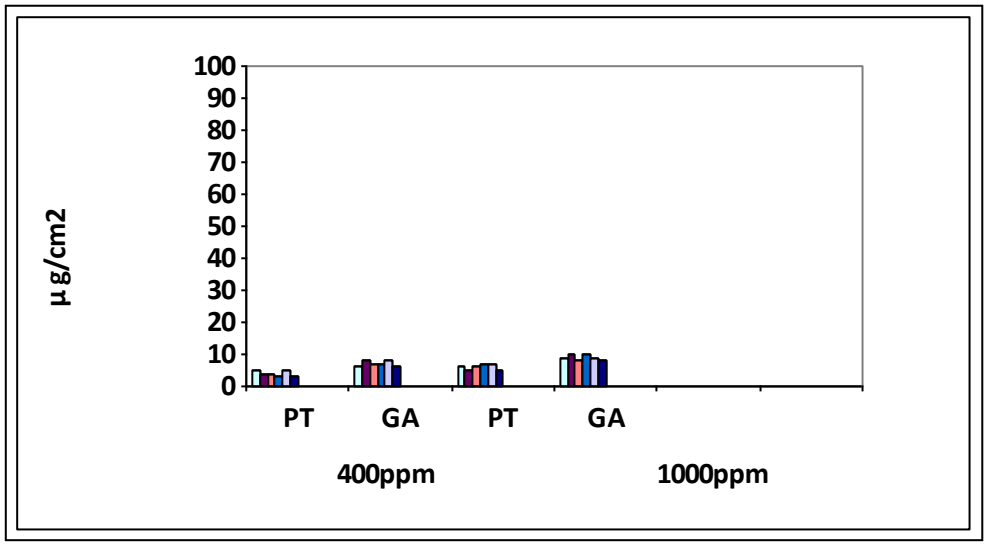



**Fig 14 Statistical analysis of elemental release graph when PT Coupled with NC alloy**

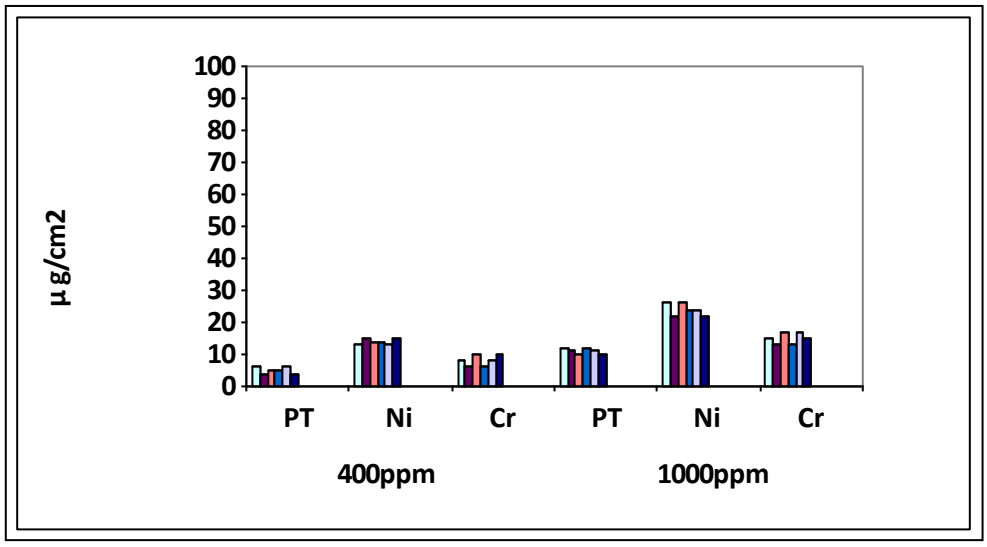


**Fig 15 Statistical analysis of elemental release graph when TA Coupled with GA**

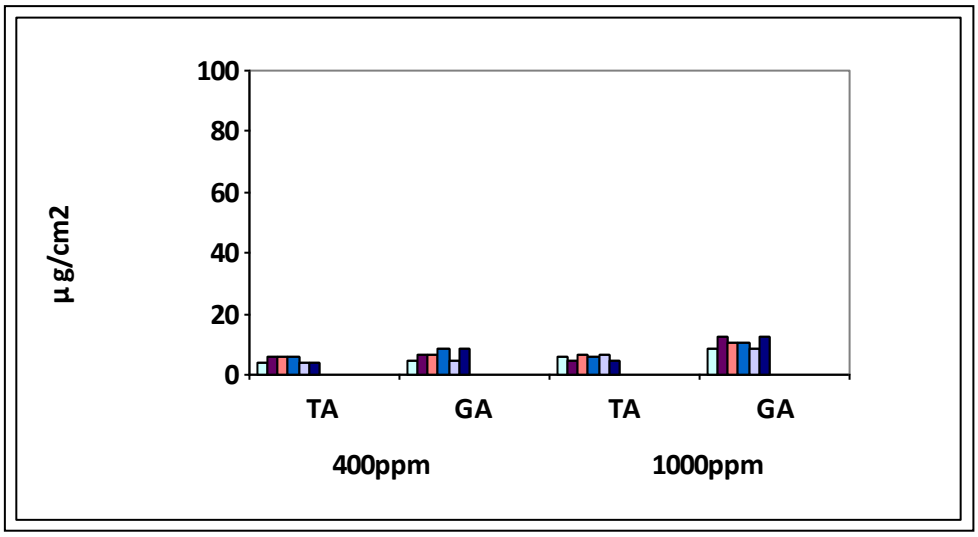
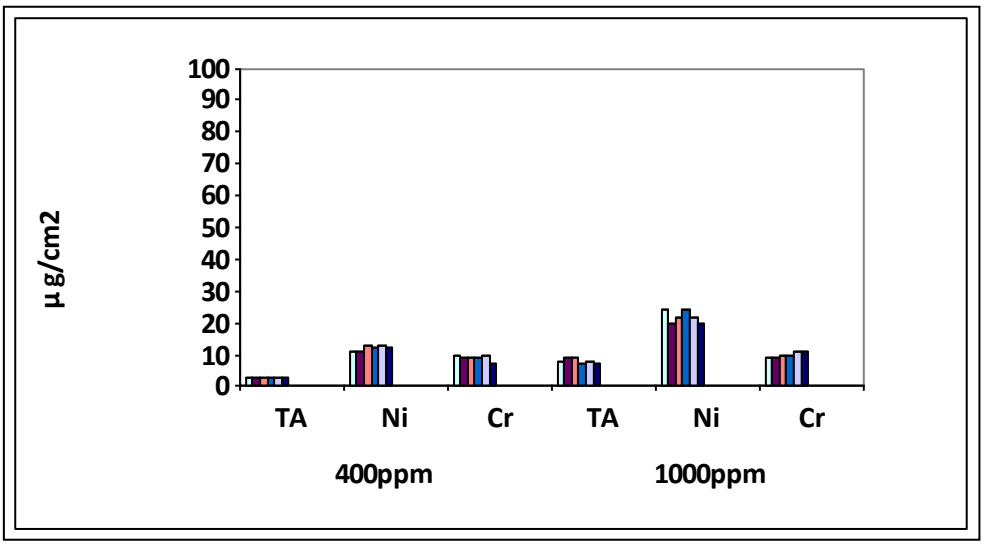



**Fig 16 Statistical analysis of elemental release graph when TA Coupled with NC alloy**



## TABLE – III

## AMOUNT OF ELEMENTS RELEASED WHEN PT AND TA COUPLED WITH GA AND NC IN μ g/ cm2

| SPECIMENS | METALS | | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | PT + GA | | | | PT + NC | | | | | | TA + GA | | | | TA + NC | | | | | |
| | 400ppm | | 000ppm | | 400ppm | | | 1000ppm | | | 400ppm | | 1000ppm | | 400ppm | | | 1000ppm | | |
| | GA | PT | GA | PT | Ni | Cr | PT | Ni | Cr | PT | GA | TA | GA | TA | Ni | Cr | TA | Ni | Cr | TA |
| 1 | 6 | 5 | 9 | 6 | 13 | 8 | 6 | 26 | 15 | 12 | 5 | 4 | 9 | 6 | 11 | 10 | 3 | 24 | 9 | 8 |
| 2 | 8 | 4 | 10 | 5 | 15 | 6 | 4 | 22 | 13 | 11 | 7 | 6 | 13 | 5 | 11 | 9 | 3 | 20 | 9 | 9 |
| 3 | 7 | 4 | 8 | 6 | 14 | 10 | 5 | 26 | 17 | 10 | 7 | 6 | 11 | 7 | 13 | 9 | 3 | 22 | 10 | 9 |
| 4 | 7 | 3 | 10 | 7 | 14 | 6 | 5 | 24 | 13 | 12 | 9 | 6 | 11 | 6 | 12 | 9 | 3 | 24 | 10 | 7 |
| 5 | 8 | 5 | 9 | 7 | 13 | 8 | 6 | 24 | 17 | 11 | 5 | 4 | 9 | 7 | 13 | 10 | 3 | 22 | 11 | 8 |
| 6 | 6 | 3 | 8 | 5 | 15 | 10 | 4 | 22 | 15 | 10 | 9 | 4 | 13 | 5 | 12 | 7 | 3 | 20 | 11 | 7 |
| Mean | 7 | 4 | 9 | 6 | 14 | 8 | 5 | 24 | 13 | 11 | 7 | 5 | 11 | 6 | 14 | 6 | 4 | 22 | 10 | 8 |

PT - Pure Titanium

TA - Titanium Alloy (Ti6Al4V)

GA - Gold Alloy

NC - Nickel Chromium Alloy

NaCl - Sodium Chloride

## STATISTICAL ANALYSIS RESULTS

Results were statistically analyzed by using one way ANOVA test. From the above results, it was found out that the pure titanium coupled with gold alloy caused negligible galvanic corrosion. There was statistically significant difference ($p < 0.001$).

# *Discussion*

# DISCUSSION

Titanium and its alloys have been considered as one of the best materials for use in dental implant applications due to their biocompatibility and high corrosion resistance mainly by the formation of titanium oxide on its surface. The nature, composition and thickness of the protective oxide scales depend on environmental conditions. Titanium has a high resistance to corrosion in physiological saline and artificial saliva. The barrier oxide was found to grow to greater thickness at lower pH and higher current density. The growth of the oxide in acid medium is lowered by an increase in temperature. The rate of dissolution of the outer layer is affected by temperature more than that of the inner layer. Saliva in the human oral cavity is generally considered to be an aerated environment.

High noble alloy in all casting protocols evaluated retained passivity under electrochemical conditions similar to the oral environment. They produced acceptable castings that are corrosion resistant when recast. Titanium is known for its high corrosion resistance due to the instant formation of an inert oxide surface layer. This has given Titanium a reputation of being a biocompatible implant material. The increasing cost of precious alloys

used in restorative dentistry has led to the development of a rapidly increasing number of low cost metallic materials for dental restorations and intraoral prostheses. Prostheses with different compositions in physical contact may develop galvanic or coupled corrosion. The anodic and cathodic parts had the same surface area, whereas in vivo, the surface areas can be considerably different, possibly modifying the intensity of the galvanic corrosion current. When a small anode is linked to a large cathode, the intensity of the corrosion process is low in the case of the Ti/dental alloys and Ti6Al4V/dental alloys[9].

In contrast, to the noble and semi-noble alloys, base metals are not as thermodynamically stable and a major aspect of their corrosion resistance is related to the formation of a thin, protective oxide layer on the surface of the metal. If the oxide layer is disrupted, then the metal or alloy must repassivate in order for the material to be protected. This stability and the ability of the passive layer to reform are important considerations when the overall corrosion properties of these alloys are being determined. Commercially pure titanium are used for dental implants and is one of the most corrosion-resistant materials used for biomedical applications. The oxide that forms on titanium provides the corrosion

resistance under static conditions and it has often been reported that titanium is not susceptible to pitting and or crevice corrosion phenomena. However, it should be pointed out that the oxide layer is not sufficiently stable to prevent galling and seizing under loading conditions. Thus, under these conditions, the titanium oxide can be removed, resulting in the release of metallic debris and ions. These properties could represent a limitations related to some dental applications[11]. The alloying elements have effect on the basic corrosion resistance of titanium in NaCl solution.

An analysis of the experimental electrochemical data and calculated parameters for each system-specific indicate that the corrosion rate of Ti-Pd is mainly controlled by anodic electron transfer reaction involved in the metal electro-dissolution process, whereas in Ti-6Al-4V, the kinetics of the electro-dissolution reaction is controlled by diffusion of OH- ions towards or the oxidation products away from the reaction sites through pores in the corrosion films[13]. The passive layers formed on titanium alloys were analysed in terms of a dual oxide layer comprising an inner barrier and an outer porous layer. The nature of the porous layer was found to depend on the nature of the alloy and the solution anion species[14]. The corrosive resistance of metal is its important characteristic

during implantation into a mouth. Therefore precious alloys are the most suitable for dental use. However, due to economic reasons, non-precious alloys (base metals) are frequently used. Because of its physical-chemical characteristics, titanium has been the material of choice during the implantation of non-precious alloys. By plating non-precious alloys with corrosive resistant alloys, the choice of implanted non-precious alloy could also spread to other economic alloys[15].

Gold alloy was an excellent couple with a titanium implant[17]. Titanium with alloy abutment and couple did not exhibit significant difference in passive current density and pitting. In the case of the gold alloy, the suprastructure showed the lowest current density. Silver–palladium alloy exhibited similar potentiodynamic polarization behavior. In the case of the cobalt–chromium alloy, the alloy exhibited the highest passive current density, the suprastructure the lowest and the couple medium :pitting occurred in all cases. The nickel–chromium alloy showed similar potentiodynamic behaviors and pitting[18]. Stress - corrosion cracking refers to cracking caused by the simultaneous presence of tensile stress and a specific corrosive medium. This process can dangerously impair the mechanical integrity of an implant. During mastication, restorations are subjected

to heavy compressive shear and bending forces. Also, burnishing of surfaces sometimes results in localized deformation. Thus, an electrolytic cell is formed between the stressed and unstressed metal portions. Grain boundaries of stressed metal are most vulnerable to corrosion. The failure of the soldered joint or the flaking away of a thin margin of gold occurs due to stress corrosion. In mouth, the process of corrosion may be of several types[20]. The elemental map showed decreased composition of Au for the recast group. The bulk composition of Au, Ag and Pd remained constant, but the copper and zinc contents slightly decreased. The results indicate that the high noble alloy tested produced acceptable castings that are corrosion resistant when recast. The high noble alloy tested in the casting protocols evaluated showed spontaneous passive behavior under electrochemical conditions similar to those in the oral environment. An artificial saliva seemed to be an acceptable medium of a corrosive environment for the high noble alloy[22].

Despite their differences of chemical compositions, most of the precious metal-based parent alloys and post-solder alloys cannot be significantly corroded when they are in contact with a neutral NaCl-containing aqueous solution. The oxidation of their constitutive elements, even of the noblest ones, is possible with significant

corrosion rates only if high values of electrical potential are applied, which cannot be really achieved by the strongest oxidant that can exist in such a solution (dissolved $O_2$).However, a galvanic corrosion can occur when a parent alloy and its usual post-solder are coupled. Fortunately, it is the alloy with the more important surface that plays the role of anode, i.e. would be corroded by galvanic coupling with very limited consequences for the mechanical resistance of the framework[24].

The electrochemical behavior of Ti-6Al-4V alloy has been studied in NaCl solutions using different techniques. From the results of open - circuit potential measurements of Ti-6Al-4V, a general tendency for the immersion potential to shift steadily towards nobler values indicated formation of a passive layer of $TiO_2$ and additional metal oxides. It was also found that Ecorr was shifted to more positive values and that Icorr increased with increasing NaCl concentration. Potentiodynamic cyclic anodic polarization measurements for Ti-6Al-4V alloy in NaCl solutions showed active, passive and trans – passive regions with the reverse scan starting below the forward scan curve, indicating that the alloy was susceptible to pitting corrosion. The effect of addition of increasing concentrations of some proteins on the corrosion of Ti-

6Al-4V alloy was also investigated using the potentiodynamic cyclic anodic polarization technique. The inhibitory action of the additives may be attributed to the formation of a thick and relatively compact film at the Ti-6Al-4V alloy surface[31]. The metallic titanium dental implant prostheses used in dentistry today derive their biocompatibility from the alloying elements responsible for the formation of a continuous stable $TiO_2$ passive film on its surface. There is a significantly small release of alloying ions even under the ideal conditions of passivity and with no damage to the implant surface. Corrosion of these implants may occur when the oral conditions are unfavorable as under mechanical trauma to the implant surface ( during placement, subject induced and trauma to assault ) or the use of inappropriate metal combination as auxiliary prostheses ( galvanism ).

The potential adverse effects of metal ion release into living tissues can be proposed based on information from literature and various clinical, preclinical and animal trial studies in-vivo and in-vitro. The results of in-vivo and invitro testing do not necessarily take into account all of the protection mechanisms and physiological host response characteristics of the actual implant environment in oral cavity. It is clear that corrosion is bound to occur and its

consequences can be quite severe[32]. A significant problem of the utilization properties forming a process of implants used in bone surgery is selection of the metallic biomaterial's mechanical properties and its physicochemical properties. The physicochemical properties of an implant's surface should be adapted to the characteristics of the human tissue environment. The corrosion resistance assessment of metallic biomaterials pertains to the basic criteria of the implant biotolerance assessment[30].

# *Conclusion*

# CONCLUSION

- PT and TA coupled with GA were more corrosion resistance compared to PT and TA coupled with NC alloy at both concentrations.
- The amount of pitting corrosion was more in NC alloy and less in GA when both were coupled with PT and TA at both concentrations.
- The amount of elemental release was more in TA compared to PT when coupled with both GA and NC alloy at both concentrations.
- The amount of elemental release was more from NC alloy when coupled with TA.
- In general among the two concentrations listed, higher concentrations significantly promoted more corrosion and among the superstructure materials listed ,GA exhibited excellent biocompatibility compared to NC alloy.
- In the galvanic corrosion study PT and GA are excellent couple compared to TA and NC alloy.
- Thus while selecting material as superstructure for titanium Implants, NC alloy must be avoided and more noble metals must be opted.

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